# HEAVY METAL CONTENT OF FLOOD SEDIMENTS AND PLANTS NEAR THE RIVER TISZA

SZILÁRD SZABÓ[1] – JÓZSEF POSTA[2] – GYÖNGYI GOSZTONYI[1] – ILONA MÉSZÁROS[3] – JÓZSEF PROKISCH[4]

[1]University of Debrecen, Department of Landscape Protection and Environmental Geography, H-4010 Debrecen, Egyetem tér 1., Hungary, E-mail: szszabo@delfin.unideb.hu; gosztonyi.gyongyi@gmail.com; [2]Department of Botany, E-mail: immeszaros@puma.unideb.hu; [4]Department of Inorganic and Analytical Chemistry, E-mail: postaj@tigris.unideb.hu; [4]University of Debrecen, Nanofood Labor, E-mail: jprokisch@agr.unideb.hu

**Abstract**
The River Tisza is Hungary's especially important river. It is significant not only because of the source of energy and the value insured by water (hydraulical power, shipping route, stock of fish, aquatic environment etc.) but the active floodplain between levees as well. Ploughlands, orchards, pastures, forests and oxbow lakes can be found here. They play a significant role in the life of the people living near the river and depend considerably on the quality of the sediments settled by the river. Several sources of pollution can be found in the catchment area of the River Tisza and some of them significantly contribute to the pollution of the river and its active floodplain. In this paper we study the concentration of zinc, copper, nickel and cobalt in sediments settled in the active floodplain and the ratio of these metals taken up by plants. Furthermore, our aim was to study the vertical distribution of these elements by the examination of soil profiles. The metal content of the studied area does not exceed the critical contamination level, except in the case of nickel, and the ratio of metals taken up by plants does not endanger the living organisms. The vertical distribution of metals in the soil is heterogeneous, depending on the ratio of pollution coming from abroad and the quality of flood.

*Keywords*: River Tisza, active floodplain, heavy metal content, plants, sediments

## 1. Introduction

Owing to the situation of Hungary in the Carpathian basin, 95 percent of the Hungarian rivers originate from beyond the frontier. Thus apart from our influential work, the water quality of the Tisza largely depends on the water coming from abroad. The object of our research is the River Tisza, which also originates from abroad, the Ukrainian Carpathians and enters Hungary at the Ukrainian-Romanian border (through Ukraine). Near the bank of the River Tisza and its tributaries, several industrial plants can be found and they constantly endanger the living organisms by neglecting the environmental directives. In the first place, mining (the barrows) and ore refining (the use of polluting technologies) cause the most significant environmental risk (Lakatos et al. 2003).

In the catchment area of the Tisza, in Romania 26, in Hungary 11, in Slovakia 1 and in Ukraine 6 hazardous potential pollution emitters can be found. Among them

there are active pollution sources as well: without any kind of treatment the sewage of many companies flows continuously into the River Szamos, the River Maros etc. and their tributaries (e. g. the liquid dung of the pig farm in Bonțida, the sewage of the paper mill in Dej, the sewage of the Therapy Pharmaceutical Factory in Cluj-Napoca), while in other cases contamination can potentially occur owing to external factors (Winkelmann-Oei et al. 2001). Some companies comply with the environmental directives, so the occurrence of an accident is less possible, and some of them do not cause pollution for the most part of the year, but a catastrophe can result in due to the unsafe ramparts. Among the former ones (i. e. the environmentally safe ones) the MOL plant in Tiszaújváros and Szolnok or the Tisza Chemical Group Public Limited Company, and among the latter ones the generally known ore refining plant of Baia Mare and Baia Borşa (accidents caused by them: 30$^{th}$ January, 2000. Baia Mare; 15$^{th}$ March, 2000. Baia Borşa) can be mentioned. These accidents occurred due to the adverse weather conditions and the intense rainfalls within a short time. In addition, contaminated slurry from mines is drained off from time to time, generally at the high-water stage of rivers and brooks so the diluted (even below the limit) contaminants get into the surface water bodies. Contaminants in water can appear in two different ways: (1) in dissolved state and (2) attached to suspended matters. Thus dissolved contaminants can become diluted, and the concentration of suspended matters is also lower in higher river flows. However, the concentration of contaminants attached to colloidal particles can be very high and can exceed the limit concerning soils and sediments, settling onto the active floodplain (Szalai, 1998, Hum and Matschullat, 2002, Braun et al. 2003, Szalai et al. 2005, Papp et al. 2008).

The traces of river pollution can be found in the sediment of the river bed or the active floodplain, and it can be proven that the contaminations in 2000 were not single cases (although they certainly resulted in high concentrations).

In this paper we examine what quantities of heavy metals can be found on the soil surface and in the deeper layers of the active floodplain in a sampling area of the Upper Tisza. Furthermore, our aim was to study what quantities of metals plants are able to intake and whether it endangers the organisms.

## 2. Materials and methods

In 2007 ten surface soil samples have been collected from the active floodplain of the River Tisza called Boroszló-kert (approximately 701 river km, near Gulács, Fig. 1). The soil samples were taken from the depth of 0-25 cm and homogenized 8-10 subsamples to make composite samples in order to decrease the errors

originated from the microheterogeneity of soil. The samples derive from ploughlands and orchards (grass among beds).

From the sampling site 5-6 stock of maize (*Zea mais*, from ploughlands) and stinging nettle (*Urtica dioica*, from orchards) was collected for metal content examinations. We chose this two plant species because we could find them everywhere in the examined area.

High resolution vertical sampling was also carried out: a 1-meter-deep soil profile was dug about 50 meters away from the mean-stage river bed inside the summer levee and sampled every two centimetres (Fig. 1, S-labelled point). In the case of sampling like this, it is very important that the area should not be disturbed (cultivated or erosion surface) because it gives rise to the mixing of the layers settled during floods, making the results unusable. So only the elder wooded areas can be taken into consideration.

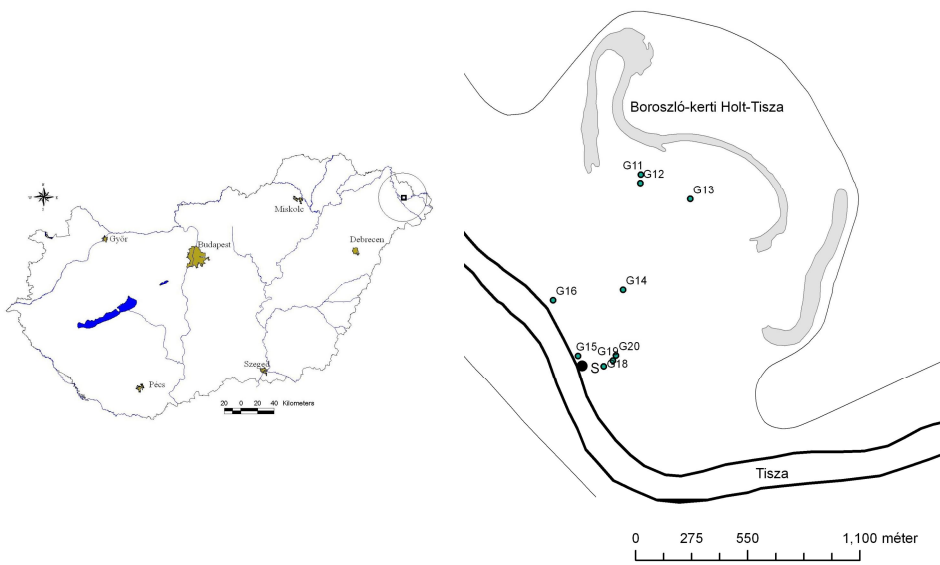


Fig. 1. The situation of the examined area and the sampling points of the Boroszlókert Dead Tisza region. Legend: S: soil profile; G11-20: surface soil samples; —: levee

The soil samples were dried at 40 °C and then passed through a 2 mm sieve. The granulometric composition (with Köhn-pipette), the humus content (after Tyurin's scheme) and the active and potential acidity ($pH_{H2O}$, $pH_{KCl}$, $y_1$, $y_2$) of soil samples were determined according to the valid Hungarian standards (MSZ-08-0210:1977, MSZ-08-0205:1978, MSZ-08-0206-2:1978). The quality of humus was carried out after Hargitai's method (1982): absorbances of 1% NaF and 0.5% NaOH extracts were measured at 533 nm (with SECOMAM spectrophotometer).

The metal content of soils has been determined according to the MSZ-08-1722-3:1989 Hungarian standard (cc. $HNO_3$+$H_2O_2$ acid digestion) with FAAS and ICP-OES. Analysis of the surface samples was carried out with Perkin-Elmer 3000 FAAS appliance (Co, Cu, Ni, Zn) at the University of Debrecen, Department of Landscape Protection and Environmental Geography. Samples from the soil profile were analysed for the same elements at the Central Chemical Laboratory of the Centre of Agricultural Sciences (University of Debrecen).

Total metal content by itself does not give enough information about the dangers caused by metals, since, depending on their form of occurrence, they are available for plants to a different extent. Therefore, in the case of surface samples the available quantity for plants has also been determined with Lakanen-Erviö extraction ($NH_4$-acetate + EDTA, Lakanen and Erviö, 1971).

The plants were washed (it was especially important in the case of roots), dried and separated into root, stem and leaves. The samples were digested (cc. $HNO_3$+$H_2O_2$), and the metal content of them was measured with an FAAS appliance.

The data were visualized by C2 (Juggins, 2003) and ArcGIS 9.0 software. In the course of data processing correlation analysis was carried out with SPSS 15.

## 3. Results and discussion

Table 1 contains the soil characteristics of the collected samples. These are little clayey, sandy-silty samples with neutral, slightly acid pH and various humus contents.

Table 1. Characteristics of soil samples (mean ± standard deviation)

| landuse | sand % | silt % | clay % | humus % | pH ($H_2O$) |
|---|---|---|---|---|---|
| ploughland | 40.0 ± 9.2 | 48.3 ± 7.4 | 11.4 ± 2.3 | 4.5 ± 1.5 | 6.84 ± 0.5 |
| orchard | 45.4 ± 9.7 | 43.5 ± 8.6 | 11.0 ± 2.6 | 5.2 ± 1.6 | 6.84 ± 0.2 |

### *3.1. Metal content of the soil samples*

The zinc contents of surface soil samples varied from 99 to 187 $mg \cdot kg^{-1}$, 140 $mg \cdot kg^{-1}$ on average. This amount is more than the background level but does not reach the critical contamination level (200 $mg \cdot kg^{-1}$, 10/2000 VI.2. KöM-EüM-FVM-KHVM directive). The amount of zinc extracted by the Lakanen-Erviö

reagent is 5-13 $mg \cdot kg^{-1}$, i. e. less than 10% of the acid-extractable concentration. The relative standard deviation of both is 18%.

The average concentration of copper is 53 $mg \cdot kg^{-1}$, the most extreme value is 80 $mg \cdot kg^{-1}$. Similarly to zinc, this amount is also beyond the background level (30 $mg \cdot kg^{-1}$) but does not exceed the contamination level. The available copper content is 8 $mg \cdot kg^{-1}$. The relative standard deviation of both extractions is approximately 25%.

The average concentration of nickel is 75 $mg \cdot kg^{-1}$, the data varies between 49 and 106 $mg \cdot kg^{-1}$. According to the 10/2000 directive, this concentration comes under the category of 'contaminated' (B value, 40 $mg \cdot kg^{-1}$). The mobilizable (Lakanen-Erviö soluble) metal content is 5 $mg \cdot kg^{-1}$ on average. The relative standard deviation is 24%. It can be seen from the available amount that the concentration is higher than the limit but it does not indicate potential danger to the biota.

In the surface soil samples, the average amount of cobalt is 20 $mg \cdot kg^{-1}$, varying between 17 and 23 $mg \cdot kg^{-1}$. The relative standard deviation of these data is the smallest (9%). The background limit is 15 $mg \cdot kg^{-1}$, the critical contamination level is 30 $mg \cdot kg^{-1}$ so the concentrations determined by us does not exceed the critical contamination level – similarly to the previous cases. The Lakanen-Erviö soluble fraction contains 3 $mg \cdot kg^{-1}$ of cobalt, and this is the only case when the relative standard deviation does not correspond with that of the acid-extractable amounts (13%).

It can be seen that, in spite of the chemical pollution sources mentioned above, the concentration of the examined metals does not exceed the critical contamination level. In our earlier work the metal content of the active floodplain and the reclaimed side was compared, and it was determined that the concentrations of the examined metals were significantly higher in the active floodplain (Szabó et al. 2008). Since these metals are essential trace elements, the higher concentration does not necessarily cause harm to the living organisms. They generally have phytotoxic effects only at very high concentrations (approximately at the double of the amount determined by us) (Hangyel, 1996, Szabó and Fodor, 1998, Naidu et al. 2003, Farsang et al. 2007, Mezősi et al. 2008.).

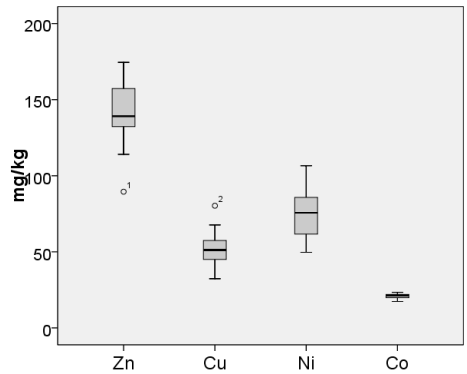


Fig. 2. The total acid-extractable metal contents of the surface soil samples

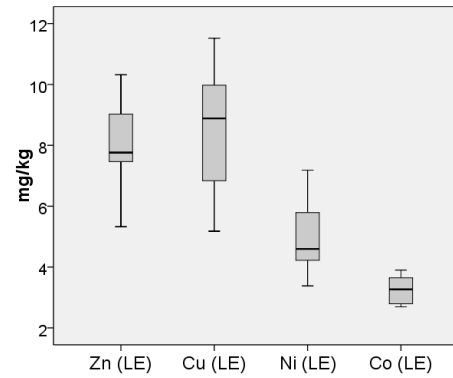


Fig. 3. The Lakanen-Erviö (LE) soluble metal contents of the surface soil samples

The standard deviation of the data refers to the heterogeneous character of the floodplain soils. The used sampling was composite sampling. It is a preferred method applied in the practical environmental protection, and its aim is to minimize the occurrence of extreme results caused by the inhomogeneity of soils (Kádár, 1998). Extremely high or low concentrations can also occur in the active floodplain, similarly to contaminated areas where large concentration differences can be found meter by meter and the effects of these extreme concentrations can only be minimized there. This can be explained by three main (closely related) reasons. (1) First, the vertical structure of soils in active floodplains is very complex due to the sediments settled by floods: depending on certain flood events, layers with different thickness, granulometric composition, humus content and pH settle and provide different conditions for the adsorption of metals. The second reason (2) is that sediments contaminated by heavy metals get regularly to the active floodplain due to the industrial activity beyond the frontier in the catchment area of the River Tisza. During the average sampling layers with different metal content (not known in advance, can be explored subsequently only by repeated sampling with large-scale uncertainty due to the microheterogeneity of soils) were taken and homogenized but – as it is apparent – great differences can be reflected even with this method. (3) As a third reason it can be mentioned that the soil is regularly ploughed in the agricultural area, which mixes the sediments in the surface soil (see above). Our results also support this: every examined soil characteristic (including metal concentrations) was significantly ($p<0.05$) different between the orchards and the ploughlands.

The diversity of vertical metal distribution is shown in Fig. 4. The metal concentrations vary in every layer and sampling area. The profile shown in the figure represents a particular instance. The intensity and the character of accumulation are different even in the nearby points of the active floodplain depending on the distance from the river, the vegetation type (roughness), the

growing season, the period of flood and the number of tributaries joined to the flow (and sediment transport) in the upper course.

In the profile (Fig. 4) the concentration of the examined metals are above the background level in the case of all metals but does not exceed the critical contamination level (10/2000 directive), only the amount of copper and zinc approaches it. Considering the endangerment of plants and – due to bioaccumulation and biomagnification – animals, it is important that the sediment settled in the past years has the highest metal content. The potentially available metal contents were measured exactly in the soil layer where the main absorbing root surface of plants can be found.

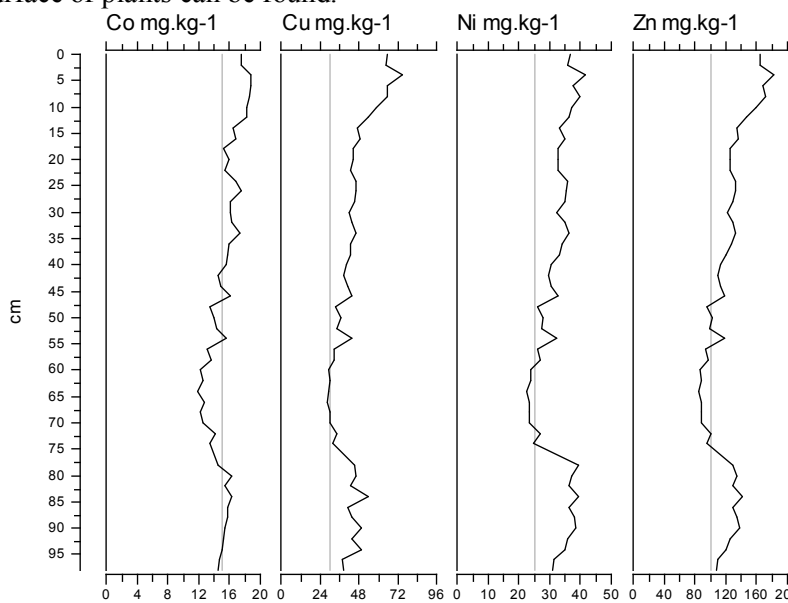


Fig. 4. The metal distribution (cobalt, copper, nickel, zinc) of the profile in the sampling site near the River Tisza (the grey line shows the background level according to the 10/2000 directive)

### *3.2. Metal content of plant samples*

Plant samples were collected and prepared in order to estimate the real risk caused by sediments with high metal content.

The zinc content of plants and plant organs (*maize* roots: 67.7 mg·kg$^{-1}$, stems: 66.3 mg·kg$^{-1}$, leaves: 16.8 mg·kg$^{-1}$; *stinging nettle* roots: 22.4 mg·kg$^{-1}$, shoots: 19.6 mg·kg$^{-1}$) does not exceed the metal content of soil (140±30 mg·kg$^{-1}$). The results show that the examined plants are not hyperaccumulator species (Fig. 5 and 6), this amount does not endanger the living organisms. In the case of maize, the crop

can even be the place of accumulation but we could not take samples from it during the sampling. The distribution of zinc is constant in the different organs of maize plants so zinc did not accumulate in the grain crop (Szabó, 2000). On the other hand it should also be noted that zinc is an essential trace element and can be harmful only at high concentrations (the lack of zinc often causes problems too). Neither the zinc content of the soil nor the concentration in the plants exceeds the health norms.

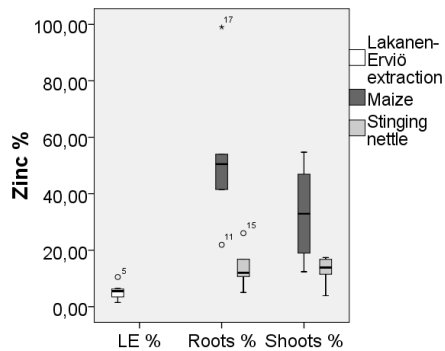


Fig. 5. The percentage of the Lakanen-Erviö soluble (LE) zinc and the zinc content in the plants compared to the total acid-extractable zinc content of soils

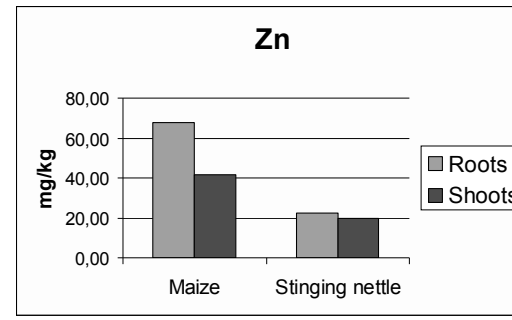


Fig. 6. The zinc concentration in the plants ($mg \cdot kg^{-1}$)

In maize the average amount of copper is higher in the roots than in the shoots (roots: 6.4 $mg \cdot kg^{-1}$, shoots: 0.9 $mg \cdot kg^{-1}$). However, the opposite can be observed in the case of stinging nettle: shoots contain more copper than the roots (roots: 0.9 $mg \cdot kg^{-1}$, shoots: 4.6 $mg \cdot kg^{-1}$). In Fig. 7 and 8 it can be seen that the amount of copper accumulated in roots and shoots on average is lower than the available metal content of soil.

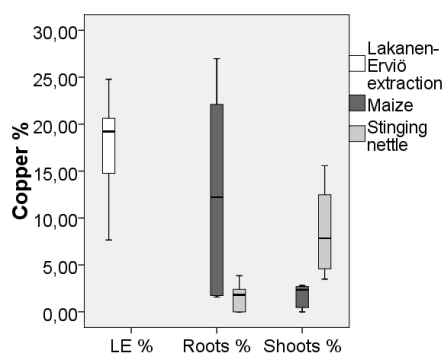


Fig. 7. The percentage of the Lakanen-Erviö soluble (LE) copper and the copper content in the plants compared to the total acid-extractable copper content of soils

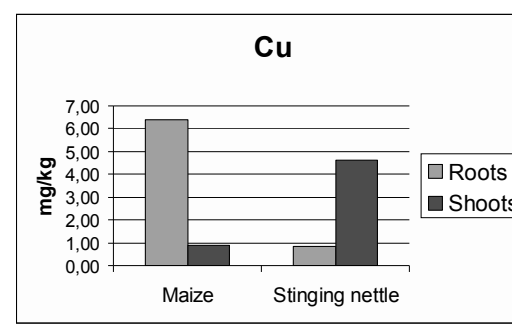


Fig. 8. The copper concentration in the plants ($mg \cdot kg^{-1}$)

Maize plants accumulated more nickel than stinging nettle (Fig. 9). However, considering the distribution of this metal the following difference can be observed in the two plants: in the case of maize more nickel is accumulated in the roots, but in stinging nettle larger quantity of nickel can be found in the shoots (Fig. 10, maize roots: 7.0 mg·kg$^{-1}$, stems: 2.4 mg·kg$^{-1}$, leaves: 2.2 mg·kg$^{-1}$; stinging nettle roots: 3.5 mg·kg$^{-1}$, shoots: 4.9 mg·kg$^{-1}$).

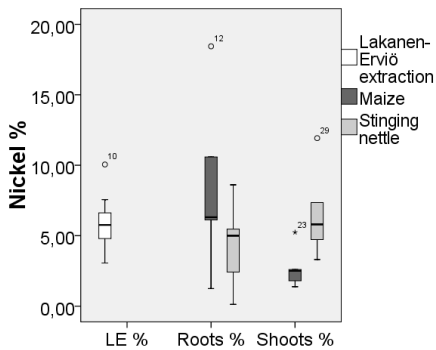


Fig. 9. The percentage of the Lakanen-Erviö soluble (LE) nickel and the nickel content in the plants compared to the total acid-extractable nickel content of soils

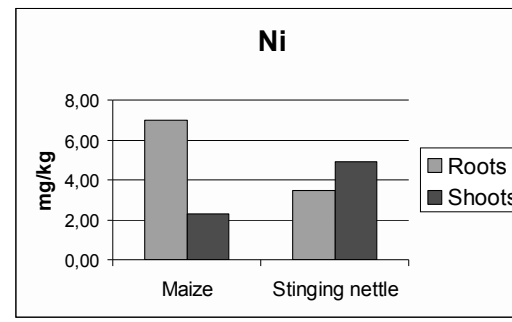


Fig. 10. The nickel concentration in the plants (mg·kg$^{-1}$)

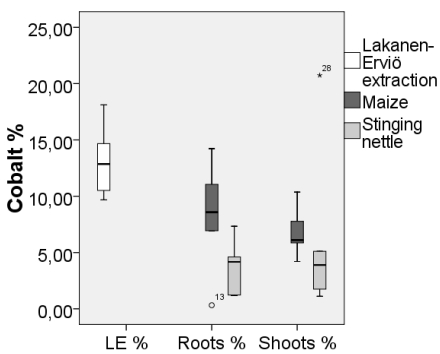


Fig. 11. The percentage of the Lakanen-Erviö soluble (LE) cobalt and the cobalt content in the plants compared to the total acid-extractable cobalt content of soils

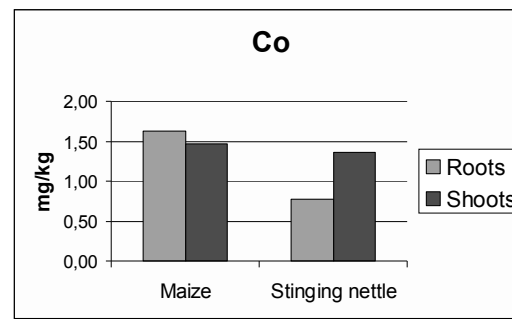


Fig. 12. The cobalt concentration in the plants (mg·kg$^{-1}$)

In the case of both maize and stinging nettle larger quantities of cobalt accumulated in the shoots. This and the amounts concentrated in the roots differ significantly from the Lakanen-Erviö soluble cobalt concentration: in the plants cobalt concentration is less than the easily soluble amount in the soil (Fig. 11-12).

On average, more cobalt accumulated in maize than in stinging nettle (maize: 1.5 $mg \cdot kg^{-1}$, stinging nettle: 1.1 $mg \cdot kg^{-1}$).

'The available element content' does not give real description of the accumulated amount: it underestimates (Zn) or overestimates (Co, Cu) the concentrations. It is taken evident since metals react differently in various solvents and, on the other hand, plants do not accumulate metals to the same extent, the strategy of metal uptake can be different in every genotype (Liu et al. 2003).

### *3.3. Correlations of soil-plant system*

Close significant correlation is observed between the soluble copper and zinc content of soils ($r=0.79$, $N=10$, $p<0.05$). It proves that copper accumulation prevents zinc uptake (Sipos 2004). Close connection can also be shown between copper concentration and humus content ($r=0.68$, $N=10$, $p<0.05$). It has been reported that humus plays a significant role in the preservation of metals and the regulation of their uptake (Livens 1991).

Close significant correlation is observed among the amount of nickel in the Lakanen-Erviö extract of soils and the plant roots and shoots. Examining the digested samples close connection was found with zinc ($r=0.64$, $N=10$, $p<0.05$). Literature proves that the presence of copper may influence the accumulation of nickel, but it is not illustrative conversely (Sipos, 2004).

During the examination of the easily mobilizable metals negative connection of cobalt with the medium-grained sand fraction appeared ($r=-0.84$, $N=10$, $p<0.05$). It can be explained by the weak adsorption on the grains of sand (Stefanovits et al. 1999).

## 4. Conclusions

Summarizing the results we can say that the metal content of the two examined plants in the active floodplain does not reach the values that limit the possibilities of utilization. Metal content changes vertically in the soil; enrichment can be observed especially in the soil surface-root zone. It may not cause any harm to plants in the region of Gulács even in the cases of zinc, copper, cobalt and nickel since they are essential trace elements. The examination of the layers from the profile helps to explain the metal content of the surface samples and the regional heterogeneity. Furthermore, the identification of the layers with extremely high or low metal concentration facilitates the age determination of them so the average

rate of sedimentation can be estimated (Kiss et al. 2004, Sándor and Kiss, 2006, Sándor and Kiss, 2007).

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